

## PATENT ABSTRACTS OF JAPAN

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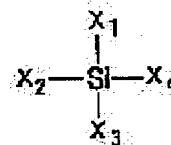
## (54) ELECTROLYTE FOR ELECTROLYTIC CAPACITOR

## (57)Abstract:

PROBLEM TO BE SOLVED: To keep high voltage-resistance for a long time by using an electrolyte containing inorganic oxide colloid particles whose surface is modified with organics.

SOLUTION: As organics, such a silylation agent or silane coupling agent as represented by equation is used

(X1-X3 are at least one kind selected among hydrocarbon group, oxyhydrocarbon group, and oxyhydroxyl group wherein, being alkyl group, alkenyl group, allyl group, or aralkyl group of carbon numbers 1-20, a part of the hydrogen may be replaced with carboxyl group, ester group, amide group, cyano group, ketone group, formyl group, ether group, hydroxyl group, amino group, mercapto group, sulfide group, sulfoxide group, sulfone group, and, X4 is alkoxy group or hydroxyl group of carbon numbers 1-20), to surface-modify inorganic oxide colloid particles. Then, the colloid particles are added to a base electrolyte by using ethylene glycol to prepare an electrolyte.



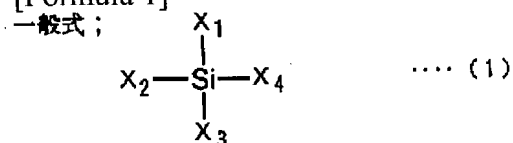
## CLAIMS

[Claim(s)]

[Claim 1] The electrolytic solution for electrolytic capacitors which consists of an inorganic oxide colloidal particle which carried out surface qualification with a solvent, a solute, and the organic substance.

[Claim 2] The electrolytic solution for electrolytic capacitors according to claim 1 whose organic substance is a sililation reagent or a silane coupling agent expressed with the following general formula (1).

[Formula 1]



A carbon number among [type X1 -X3 The alkyl group of 1-20, an alkenyl radical, They are an aryl group or an aralkyl radical. A part of the hydrogen A carboxyl group, An ester group, an amide group, a cyano group, a ketone group, a formyl group, a ether group, it was chosen out of the group of a hydroxyl group, the amino group, a sulfhydryl group, a sulfide radical, a sulfoxide radical, the hydrocarbon group (- R) that may be replaced by the sulfone radical, an oxy-hydrocarbon group (-OR), and a hydroxyl group (- OH) -- it is a kind at least and you may differ mutually. X4 It is the alkoxy group or hydroxyl group of \*\*\*\*\* 1-20.]

[Claim 3] The electrolytic solution for electrolytic capacitors according to claim 2 whose sililation reagent or silane coupling agent expressed with a general formula (1) is one or more sorts chosen from 3-glycidoxypopyltrimetoxysilane, 3-glycidoxypopyltriethoxysilane, 3-glycidoxypopylmethyldimethoxysilane, and 3-glycidoxypopylmethyldiethoxysilane.

[Claim 4] The electrolytic solution for electrolytic capacitors according to claim 1 whose inorganic oxide colloidal particle is a silica.

[Claim 5] The electrolytic solution for electrolytic capacitors according to claim 4 characterized by particle size of a silica being 5-100nm.

[Claim 6] The electrolytic solution for electrolytic capacitors according to claim 1 which is the organic solvent with which a solvent makes ethylene glycol or gamma-butyrolactone a subject.

[Claim 7] The electrolytic solution for electrolytic capacitors according to claim 1 whose solute is the onium salt of an organic acid and/or an inorganic acid.

[Claim 8] The electrolytic solution for electrolytic capacitors according to claim 7 whose organic acid is one or more sorts chosen from 1, 6-Deccan dicarboxylic acid, 1, 7-octane dicarboxylic acid, an adipic acid, a benzoic acid, a phthalic acid, and a maleic acid.

[Claim 9] The electrolytic solution for electrolytic capacitors according to claim 7 whose inorganic acid is a boric acid.

[Claim 10] The electrolytic solution for electrolytic capacitors according to claim 7 which is one or more sorts chosen from a group which an onium salt becomes from ammonium salt, the third class ammonium salt, quarternary ammonium salt, and a friend JINIUMU

salt.

[Claim 11] The electrolytic solution for electrolytic capacitors according to claim 1 which is the silica particle in which a solvent with which a solvent makes ethylene glycol a subject, and an inorganic oxide colloidal particle a solute carried out [ a colloidal particle ] surface qualification with ammonium salt of an organic acid and the organic substance carried out surface treatment by silane coupling agent.

[Claim 12] The electrolytic solution for electrolytic capacitors according to claim 1 which is the silica particle in which a solvent with which a solvent makes gamma-butyrolactone a subject, and an inorganic oxide colloidal particle a solute carried out [ a colloidal particle ] surface qualification with the third class ammonium salt of an organic acid, quarternary ammonium salt or a friend JINIUMU salt, and the organic substance carried out surface treatment by silane coupling agent.

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[Translation done.]

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## DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] This invention relates to the electrolytic solution for electrolytic capacitors. Furthermore, it is related with the electrolytic solution for electrolytic capacitors which can offer the electrolytic capacitor which has high withstand voltage and high reliability by making the inorganic oxide colloidal particle which carried out surface qualification with the organic substance in detail contain.

[0002]

[Description of the Prior Art] An electrolytic capacitor uses for an anode plate the so-called valve metal with which insulating oxide film layers, such as aluminum and a tantalum, may be formed, and uses for an anode plate lateral electrode what formed the aforementioned insulating oxide film thin film for the surface as a dielectric layer by anodizing etc. And generally winding mold element structure which is illustrated by drawing 1 as this example is known, and make an anode plate lateral electrode (1) counter, arrange a cathode lateral electrode (2), a separator (3) is made to intervene between an anode plate lateral electrode and a cathode lateral electrode, and the electrolytic solution is made to hold to this separator. It puts into the sheathing case (5) of the quality of the materials, such as aluminum as shows this to drawing 2, and has structure which sealed this case using obturation boards (7), such as a phenolic laminated sheet, polypropylene, and a polyphenylene sulfide, through rubber packing (6), such as isobutylene isoprene rubber, ethylene propylene rubber, and silicone rubber.

[0003] In the aluminium electrolytic condenser which used the aluminum oxide for the dielectric, etching processing of the anode plate lateral electrode is usually carried out for expansion of surface area. The electrolytic solution is close to the concavo-convex side of this anode plate lateral electrode, and functions as substantial cathode which transmits the electric field of a cathode lateral electrode. For this reason, the conductivity of the electrolytic solution, the temperature characteristic, etc. are the factor which determines the electrical characteristics [an impedance, dielectric loss ( $\tan\delta$ ), equivalent series

resistance (ESR), etc.] as an electrolytic capacitor. Moreover, the role (formation sex) which restores the insulating deterioration and the damage on an oxide film thin film is required of the electrolytic solution, and this affects it to the leakage current (LC) and the life property of an electrolytic capacitor. Thus, the electrolytic solution is an important component which influences the property of an electrolytic capacitor.

[0004] The conductivity of the electrolytic solution has the desirable electrolytic solution which has high conductivity from being directly concerned with the energy loss of an electrolytic capacitor, an impedance characteristic, etc. The electrolytic capacitor which has higher withstand voltage so that neither short-circuit nor ignition may be started from a rise of the demand to safety on the other hand under a severe condition to which the abnormal voltage which exceeds rated voltage to an electrolytic capacitor is impressed is called for. However, if the conductivity of the electrolytic solution to be used generally becomes high, the withstand voltage of an electrolytic capacitor tends to fall and makes development of an electrolytic capacitor difficult (Ue et al., a new capacitor, three volumes, 55 pages, 1996). Then, adding various inorganic oxide colloidal particles to the electrolytic solution as an electrolytic capacitor \*\*\*\* attempt in which it has high withstand voltage, and raising withstand voltage is examined, using the electrolytic solution which has high conductivity.

[0005] For example, raising withstand voltage is proposed by adding a silica colloidal particle to the electrolytic solution, maintaining the high conductivity of the electrolytic solution (JP,1-232713,A). Moreover, adding an alumina (JP,4-145612,A), a zirconia (JP,4-145613,A), a titania (JP,4-145616,A), aluminosilicate (JP,6-283388,A), an aluminosilicate covering silica (JP,6-349684,A), etc. besides a silica is also proposed.

[0006]

[Problem(s) to be Solved by the Invention] However, in the electrolytic solution containing these inorganic oxide colloidal particle, although early withstand voltage was high, withstand voltage fell during life test and there was a trouble that short-circuit occurred. Moreover, in the case of the electrolytic solution which used the ion of many \*\*, such as dicarboxylic acid, as a solute, especially this withstand voltage fall phenomenon was remarkable.

[0007]

[Means for Solving the Problem] As a phenomenon common to the electrolytic solution with which such withstand voltage falls, gelation of the electrolytic solution and precipitate generation of an inorganic oxide were checked during life test, and in order to carry out long duration continuation of the improvement effect in withstand voltage, that an added inorganic oxide colloidal particle maintains a stable colloidal state, without causing gelation and precipitate found out that there was necessity. For the above-mentioned purpose, as a result of inquiring wholeheartedly, an inorganic oxide colloidal particle which carried out surface qualification with the organic substance completed a header and this invention very for it being hard to cause gelation and precipitate in the electrolytic solution for capacitors compared with the conventional inorganic oxide colloidal particle. That is, this invention relates to the electrolytic solution for electrolytic capacitors characterized by realizing an electrolytic capacitor which can carry out long duration maintenance of the high withstand voltage at an elevated temperature by using the electrolytic solution containing an inorganic oxide colloidal particle which carried out surface qualification with the organic substance.

[0008]

[Function] By carrying out surface qualification of the inorganic oxide colloidal particle with the organic substance, compatibility with an organic solvent improves, condensation of a particle is barred, and even if it carries out long duration neglect at an elevated temperature, neither gelation nor precipitate is produced. Therefore, it is possible to carry out long duration continuation of the improvement effect in withstand voltage of the inorganic oxide colloidal particle in an electrolytic capacitor.

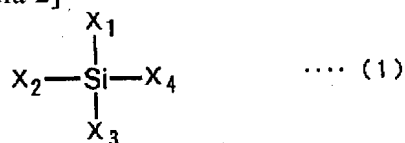
[0009]

[Embodiment of the Invention] Hereafter, this invention is explained to details. As the organic substance which carries out surface qualification of the inorganic oxide colloidal particle used by this invention, various high molecular compounds, such as a sililation reagent, a silane coupling agent, a titanate system coupling agent, an aluminate coupling agent, alcohols, and a latex, etc. can be mentioned.

[0010] A sililation reagent and a silane coupling agent are expressed with the following general formula (1).

[Formula 2]

一般式;



A carbon number among [type X1 -X3 The alkyl group of 1-20, an alkenyl radical, They are an aryl group or an aralkyl radical. A part of the hydrogen A carboxyl group, An ester group, an amide group, a cyano group, a ketone group, a formyl group, a ether group, it was chosen out of the group of a hydroxyl group, the amino group, a sulfhydryl group, a sulfide radical, a sulfoxide radical, the hydrocarbon group (- R) that may be replaced by the sulfone radical, an oxy-hydrocarbon group (-OR), and a hydroxyl group (- OH) -- it is a kind at least and you may differ mutually. X4 It is the alkoxy group or hydroxyl group of \*\*\*\*\* 1-20.]

[0011] X1 - -- X3 As an example, an oxy-hydrocarbon group or hydroxyl groups, such as hydrocarbon groups, such as aralkyl radicals, such as aryl group; benzyls, such as alkenyl radical; phenyl groups, such as alkyl group; vinyl groups, such as a methyl group, an ethyl group, a propyl group, butyl, a decyl group, and an octadecyl radical, and an allyl group, and a naphthyl group, and a phenethyl radical, a methoxy group, an ethoxy radical, a propoxy group, a butoxy radical, a vinyloxy radical, a phenoxy group, and a benzyloxy radical, can be mentioned Furthermore, sulfhydryl groups, such as amino-groups; 3-mercapto propyl groups, such as epoxy group; 3-aminopropyl radicals, such as acrylic radical; 3-glycidoxy propyl groups, such as 3-methacryloxypropyl radical, and 2-(3, 4-epoxycyclohexyl) ethyl group, an N-phenyl-3-aminopropyl radical, and an N-(2-aminoethyl)-3-aminopropyl radical, can be mentioned as an example in the case of having a substituent. X4 As \*\*\*\*\* , alkoxy group; hydroxyl groups, such as a methoxy group, an ethoxy radical, a propoxy group, and a butoxy radical, can be mentioned.

[0012] Also in such combination, methyl trimetoxysilane, methyl triethoxysilane, Dimethyl dimethoxysilane, dimethyl diethoxysilane, phenyl trimethoxysilane, Phenyl triethoxysilane, diphenyl dimethoxysilane, diphenyl diethoxysilane, Isobutyl trimethoxysilane, isobutyl triethoxysilane, decyltrimetoxysilane, Decyltriethoxysilane,

vinyltrimethoxysilane, vinyltriethoxysilane, 3-methacryloxy propyl trimethoxysilane, 3-methacryloxypropyl triethoxysilane, 3-glycidoxypropyltrimethoxysilane, 3-glycidoxypropyltriethoxysilane, 3-glycidoxypropylmethyldimethoxysilane, 3-glycidoxypropyl methyldiethoxysilane, 2-(3, 4-epoxycyclohexyl) ethyl TORITORI methoxy silane, 2-(3, 4-epoxycyclohexyl) ethyl TORITORI ethoxy silane, 3-aminopropyl trimethoxysilane, 3-aminopropyl triethoxysilane, N-phenyl-3-aminopropyl trimethoxysilane, N-phenyl-3-aminopropyl triethoxysilane, N-(2-aminoethyl)-3-aminopropyl trimethoxysilane, N-(2-aminoethyl)-3-aminopropyl triethoxysilane, 3-mercapto propyltrimethoxysilane, 3-mercapto propyl triethoxysilane, etc. are desirable. The 3-glycidoxypropyltrimethoxysilane which has 3-glycidoxy propyl group with sufficient solvents, such as ethylene glycol and gamma-butyrolactone, and compatibility also in it, 3-glycidoxypropyltriethoxysilane, 3-glycidoxypropylmethyldimethoxysilane, and especially 3-glycidoxy propyl methyldiethoxysilane are desirable.

[0013] As an example of a titanate system coupling agent Isopropylisostearoyl titanate, isopropyl tridodecyl benzenesulphonyl titanate, Isopropyl tris (dioctyl pyrophosphate) titanate, tetra-isopropyl screw (dioctyl phosphite) titanate, Tetra-octyl screw (JITORIDE sill phosphite) titanate, tetrapod (2 and 2-diaryl oxymethyl-1-butyl) screw (JITORIDESHIRU) phosphite titanate, Screw (dioctyl pyrophosphate) oxy-acetate titanate, Isopropanal PIRUTORI octanoyl titanate, isopropyl JIMETAKU roil isostearoyl titanate, Isopropanal PIRUTORI (dioctyl phosphate) titanate, isopropyl TORIKUMIRU phenyl titanate, isopropanal PIRUTORI (N-aminoethyl aminoethyl) titanate, etc. are mentioned.

[0014] As an example of an aluminate coupling agent, aluminum ethylacetoacetate diisopropylate, aluminum tris (ethyl acetoacetate), aluminum tris (acetylacetonate), aluminum screw (ethyl acetoacetate) monoacetyl acetonate, etc. are mentioned. As an example of alcohol, a methanol, ethanol, n-propanol, iso-propanol, n-butanol, amyl alcohol, 4-methyl-2-pentanol, n-heptanol, n-octanol, 2-ethylhexanol, nonanol, decanol, tridecanol, 2-methoxyethanol, 2-ethoxyethanol, 2-butoxyethanol, 3-methoxybutanol, 3-methyl-3-methoxybutanol, polyvinyl alcohol, etc. are mentioned.

[0015] The organic substance used for surface qualification of these sililation reagents, a silane coupling agent, a titanate system coupling agent, an aluminate coupling agent, alcohols, various high molecular compounds, etc. is independent, or can be used in two or more combination. As an example of the inorganic oxide colloidal particle used by this invention, such mixture, such as a silica, an alumina, a titania, a zirconia, antimony oxide, aluminosilicate, a silica zirconia, a titania zirconia, an aluminosilicate covering silica, and a silica zirconia covering silica, is mentioned. A silica, aluminosilicate, and an aluminosilicate covering silica are desirable especially especially from a viewpoint of the ease of silanizing processing, the stability of colloid, and the improvement effect of withstand voltage.

[0016] The range of the mean particle diameter of an inorganic oxide colloidal particle is 5-100nm preferably, and the range of it is 10-50nm still more preferably. If the particle size of an inorganic oxide colloidal particle is too small, gelation of the electrolytic solution tends to take place, and if too large, it will be easy to produce precipitate, and will be hard to become stable colloid. The inorganic oxide colloidal particle which carried out surface qualification by the sililation reagent or silane coupling agent used by this invention can be obtained by the method of a publication for example, on U.S. Pat.

No. 4,027,073 specifications. The inorganic oxide colloidal particle which carried out surface qualification in the alcohol used by this invention can be obtained by the method of a publication for example, on U.S. Pat. No. 2,657,149 specifications.

[0017] Especially as the addition method of the inorganic oxide colloidal particle which carried out surface qualification with these organic substance, although not limited, since these hardly melt into a solvent, the method of adding to the electrolytic solution as the colloidal solution which the suitable dispersion medium was made to distribute generally is desirable. Although there is especially no limitation as a dispersion medium, if the ethylene glycol which is the aforementioned solvent is used here, there is also little effect of [ on the property to the basic electrolytic solution ], and, moreover, diffusion into the electrolytic solution is also easy for it. The addition of this inorganic oxide colloidal particle is 0.5 - 18% of the weight of the electrolytic solution preferably, and especially a desirable thing is 6 - 10% of the weight of a range. If many [ if there are too few additions of an inorganic oxide colloidal particle, a withstand voltage rise of the electrolytic solution is not enough, and / too ], it will be easy to produce gelation and precipitate and will be hard to become stable colloid.

[0018] In addition, although the silane coupling agent may react chemically with a solvent, a solute, etc. which were used after it performs qualification processing in case it performs qualification processing to an inorganic oxide colloidal particle or since it has the reactant functional group, it does not pose a problem especially in this invention. For example, although alcoholysis is carried out and hydrolysis and carrying out ring breakage have an epoxy group when 3-glycidoxypropyltrimetoxysilane is used as a silane coupling agent, it is not a \*\*\*\*\* thing about a bad influence at all.

[0019] As an example of the solvent used by this invention, ethylene glycol, a glycerol, Alcoholic solvents, such as methyl cellosolve; Gamma-butyrolactone, gamma-valerolactone, Lactone solvents, such as delta-valerolactone; N-methyl formamide, N-ethyl formamide, N,N-dimethylformamide, N,N-dimethylacetamide, Amide solvents, such as N-methyl pyrrolidinone; Ethylene carbonate, Nitril solvents, such as carbonate solvent; 3-methoxy propionitriles, such as propylene carbonate and butylene carbonate, and guru taro nitril; such mixture [ , such as a phosphoric ester solvent, ], such as trimethyl phosphate and phosphoric-acid triethyl, is mentioned. The ethylene glycol and gamma-butyrolactone which are the organic solvent with which the electrolytic solution which has big solvent power to various kinds of solutes especially, and was excellent in the temperature characteristic is obtained are desirable.

[0020] As an example of the organic-acid component used in the onium salt of the organic acid used as a solute, and/or an inorganic acid by this invention A benzoic acid, a toluic acid, a cumin acid, t-butyl benzoic acid, a salicylic acid, Aromatic series monocarboxylic acid, such as an anisic acid; A formic acid, an acetic acid, a propionic acid, Aliphatic series monocarboxylic acid, such as a 7-phenyl-7-methoxy-1-octane carboxylic acid and a 6-phenyl-6-methoxy-1-heptane carboxylic acid; A phthalic acid, Aromatic series dicarboxylic acid, such as 4-methyl phthalic acid and 4-nitro phthalic acid; A maleic acid, Partial saturation aliphatic series dicarboxylic acid, such as citraconic-acid, dimethyl maleic-acid, 1, and 2-cyclohexene dicarboxylic acid; Oxalic acid, A malonic acid, a succinic acid, a glutaric acid, an adipic acid, a pimelic acid, a suberic acid, Straight chain-like saturation aliphatic series dicarboxylic acid, such as an azelaic acid, a sebacic acid, undecane diacid, dodecane diacid, and a tridecanedioic acid;

A dimethyl malonic acid, A diethyl malonic acid, a dipropyl malonic acid, 2-methyl glutaric acid, 3-methyl glutaric acid, 3 and 3-dimethyl glutaric acid, 3-methyl adipic acid, 2 and 2, a 4-trimethyl adipic acid, A 2, 4, and 4-trimethyl adipic acid, 1, 6-Deccan dicarboxylic acid, 5, 6-Deccan dicarboxylic acid, 1, 7-octane dicarboxylic acid, the 7-methyl-7-KARUBO methoxy -1, 9-Deccan dicarboxylic acid, 2, 8-nonane dicarboxylic acid, 7, 8 and 11, 12-tetramethyl - 1, 18-OKUTA decane dicarboxylic acid, 1-methyl-3-ethyl -1, 7-heptane dicarboxylic acid, 1, the 3-dimethyl -1, 7-heptane dicarboxylic acid, The 5-methyl -1, 7-octane dicarboxylic acid, 7, the 12-dimethyl -1, 18-OKUTA decane dicarboxylic acid, 7-ethyl -1, 16-hexadecane dicarboxylic acid, 7, the 8-dimethyl -1, 14-tetradecane dicarboxylic acid, 1, 6-heptane dicarboxylic acid, the 6-methyl-6-KARUBO methoxy -1, 8-nonane dicarboxylic acid, 1, 8-nonane dicarboxylic acid, the 8-methyl-8-KARUBO methoxy -1, 10-undecane dicarboxylic acid, Saturation aliphatic series dicarboxylic acid; 7-methyl which has the branched chain of 6-ethyl -1, 4-tetradecane dicarboxylic acid, cyclohexane dicarboxylic acid, etc. - 1, 7, 9-Deccan tricarboxylic acid, 6-methyl - 1, 6, 8-nonane tricarboxylic acid, 8-methyl - Such mixture [ , such as tricarboxylic acid, ], such as 1, 8, and 10-undecane tricarboxylic acid, is mentioned. Moreover, a boric acid, phosphoric acid, etc. are mentioned as an example of an inorganic-acid component.

[0021] The phthalic acid from which the electrolytic solution with high conductivity is obtained also among the above-mentioned organic-acid component and an inorganic-acid component for [ not more than rated voltage 100V / for low voltage ] capacitors, a maleic acid, a benzoic acid, and an adipic acid are desirable. The azelaic acid and sebacic acid with which the electrolytic solution with high withstand voltage is obtained for [ beyond rated voltage 300V / for high pressures ] capacitors, 1, 6-Deccan dicarboxylic acid, 1, 7-octane dicarboxylic acid, and a boric acid are desirable. Rated voltage 100V are exceeded and the benzoic acid with which the electrolytic solution which has the conductivity and withstand voltage moderate for [ for medium voltages ] capacitors below 300V is obtained, an adipic acid, and an azelaic acid are desirable.

[0022] as the example of an onium salt -- 3, such as ammonium; methylammonium; dimethylammonium; trimethylammonium, ethyl dimethylammonium, diethyl methylammonium, and triethyl ammonium, -- 4, such as class ammonium; tetramethylammonium, triethyl methylammonium, and tetraethylammonium, -- such mixture [ , such as friend JINIUMU, ], such as class ammonium; 1, 2 and 3, 4-tetramethyl imidazolinium, 1-ethyl -2, and 3-dimethyl imidazolinium, is mentioned.

[0023] To the capacitor for inside high pressures, the ammonia with which the electrolytic solution which has high withstand voltage in the combination of an ethylene glycol solvent and dicarboxylic acid, such as 1 and 6-Deccan dicarboxylic acid, is obtained is desirable. To the capacitor for low voltage, 1, 2 and 3 from which a gamma-butyrolactone solvent and the electrolytic solution which has high conductivity in combination, such as a phthalic acid, are obtained, 4-tetramethyl imidazolinium, 1-ethyl -2, 3-dimethyl imidazolinium, tetramethylammonium, triethyl methylammonium, and tetraethylammonium are desirable. As for the amount of the solute used, it is desirable to make it contain in 5 - 30% of the weight of the range to the sum total weight of a solvent and a solute.

[0024] moreover, this invention -- setting -- formation -- the electrolytic solution can also be made to contain water for the purpose, such as improvement in a sex The content of



this water is 0.01 - 30% of the weight of a range preferably, and is 0.01 - 10% of the weight of a range still more preferably. Moreover, the electrolytic solution can also be made to contain the additive of further others if needed. as other additives, nitro compounds, such as phosphorus-compounds; p-nitrobenzoic acid of boron compound; phosphoric acids, such as complex compounds (ethylene glycol, a mannitol, sorbitol, etc.) of a boric acid, a boric acid, and polyhydric alcohol, alkyl acid phosphate [phosphoric-acid dibutyl and a phosphoric-acid screw (2-ethylhexyl)], and acid phosphonate [2-ethylhexyl phosphonic acid (2-ethylhexyl) etc.] and m-nitro acetophenone, are mentioned. The electrolytic solution of this invention can be used for the aluminium electrolytic capacitor of the winding mold shown in drawing 1 and drawing 2, and impregnation of this electrolytic solution is carried out to the separator (it is also called a spacer) shown by 3 all over drawing. Generally as for this separator, kraft paper, Manila paper, etc. are used.

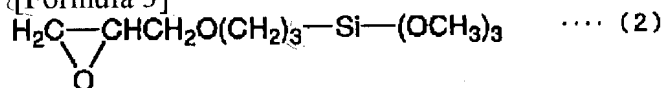
[0025]

[Example] Hereafter, although this invention is concretely explained based on an example, this invention is not limited at all by these examples. The conductivity and withstand voltage in 25 degrees C of the electrolytic solution using the usual silica colloidal particle which has not carried out surface qualification with the organic substance were shown in the electrolytic solution of this invention using the silica colloidal particle which carried out surface qualification with the organic substance, and a table 2 (1 [ the ], its 2) at a table 1 (1 [ the ], its 2), respectively.

[0026] Among the table, unless it refused, the weight section when setting a solvent and the sum total of a solute to 100 showed especially the amount used and the addition of each component. Remaining as it is (example of a comparison) or the thing (example) which carried out surface qualification using the 3-glycidoxypropyltrimetoxysilane expressed with the following general formula (2) was used for the inorganic oxide colloidal particle for the silica particle whose mean particle diameter is about 12nm. Using ethylene glycol as a dispersion medium of colloid, colloid was added to the basic electrolytic solution (a solute and solvent), and the electrolytic solution of a predetermined presentation was prepared.

[0027]

[Formula 3]



[0028] Withstand voltage sank into the winding mold element which showed these electrolytic solutions to drawing 1, and was made into the voltage value by which a spike or scintillation was first observed by the ascending curve of the voltage-time amount when impressing constant current to this. The specification of the aluminium electrolytic condenser element used in examples 1-4 and the examples 1-4 of a comparison is a thing with a rated voltage 450V and a rated electrostatic capacity of 10 micro F. The specification of the aluminium electrolytic condenser element used in examples 5-11 and the examples 5-11 of a comparison is a thing with a rated voltage 200V and a rated electrostatic capacity of 68 micro F. The current values impressed by measurement of withstand voltage are 3mA and 10mA, respectively. The experimental result of a table 1 and a table 2 shows that the electrolytic solution of this invention has conductivity

equivalent to the conventional electrolytic solution, and higher withstand voltage.

[0029]

[A table 1]

表 1

	電解液組成（重量部）								電導度 (mS/cm)
	溶 質		溶 媒		添 加 劑				
	a ~ k	部	B	C	D	E	F	G	
実施例 1	a	15	85		2	6	—	—	1.79
実施例 2	b	15	85		2	6	—	—	1.71
実施例 3	c	10	90		2	6	—	—	3.17
実施例 4	d	10	90		2	6	—	—	2.39
実施例 5	e	20	16	64	2	6	—	—	8.50
実施例 6	f	20	16	64	2	6	—	—	9.30
実施例 7	g	20	16	64	2	6	—	—	11.37
実施例 8	h	20	16	64	2	6	—	—	6.84
実施例 9	h	20	16	64	2	6	—	2	6.44
実施例 10	i	20	16	64	2	6	—	2	7.48
実施例 11	j	20	16	64	2	6	—	—	7.73
実施例 12	k	—	—	—	—	6	—	—	0.95

## 溶 質

- a : 1, 6-デカジカルボン酸アンモニウム  
 b : 1, 7-オクタンジカルボン酸アンモニウム  
 c : アジピン酸アンモニウム  
 d : 安息香酸アンモニウム  
 e : フタル酸水素 1-エチル-2, 3-ジメチルイミダゾリ  
 f : フタル酸水素テトラメチルアンモニウム  
 g : マレイン酸水素トリエチルメチルアンモニウム  
 h : 安息香酸トリエチルメチルアンモニウム  
 i : 安息香酸 1-エチル-2, 3-ジメチルイミダゾリ-9

[0030]

[A table 2]

表 2

	電解液組成（重量部）								電導度 (mS/cm)	耐電圧 (V)
	溶 質		溶 媒		添 加 剤					
	a ~ k 部	B	C	D	E	F	G			
比較例 1	a	15	85	—	2	—	6	—	1.74	590
比較例 2	b	15	85	—	2	—	6	—	1.70	530
比較例 3	c	10	90	—	2	—	6	—	3.14	585
比較例 4	d	10	90	—	2	—	6	—	2.36	520
比較例 5	e	20	16	64	2	—	6	—	8.31	125
比較例 6	f	20	16	64	2	—	6	—	8.99	100
比較例 7	g	20	16	64	2	—	6	—	11.15	145
比較例 8	h	20	16	64	2	—	6	—	6.74	175
比較例 9	h	20	16	64	2	—	6	2	6.34	220
比較例 10	i	20	16	64	2	—	6	2	7.34	> 260
比較例 11	j	20	16	64	2	—	6	—	7.66	175
比較例 12	k	—	—	—	—	—	6	—	0.94	575

溶質、a ~ k、溶媒 B、C、添加剤 D ~ G は表 1 に記載の通りである。

[0031]

[A table 3]

表 3

電解液	初期値		高温負荷試験	
	静電容量 ( $\mu$ F)	$\tan \delta$	静電容量 変化率(%)	$\tan \delta$
実施例 1	11.0	0.040	-1.1	0.044
実施例 2	11.1	0.041	-1.0	0.044
比較例 1	11.0	0.040	ショート (10 個中、7 個)	
比較例 2	11.1	0.041	ショート (10 個中、7 個)	

[0032]

[A table 4]

表 4

電解液	ゲル化までの日数 (115℃)
実施例 1	22日
実施例 2	18日
実施例 3	5日
実施例 4	30日以上
実施例 5	
実施例 6	
実施例 7	
実施例 8	
実施例 9	
実施例 10	
実施例 11	
比較例 1	1日
比較例 2	1日
比較例 3	1日
比較例 4	1日
比較例 5	1日
比較例 6	1日
比較例 7	1日
比較例 8	2日
比較例 9	2日
比較例 10	1日
比較例 11	1日

[0033] The electrolytic solution of the examples 1 and 2 of this invention and the conventional examples 1 and 2 is used for a table 3, rated voltage 450V and an aluminium electrolytic condenser with an electrostatic capacity of 10 micro F are produced, and the result at the time of performing the elevated-temperature load test of 1000 hours at 105 degrees C is shown. As for the electrolytic capacitor of this invention, after the elevated-temperature load test maintained initial value so that the experimental result of a table 3 might show. On the other hand, much short articles were generated during the elevated-temperature load test by the fall of withstand voltage, and the conventional electrolytic capacitor had a problem in reliability. These electrolytic solutions were sealed in the vial pipe to a table 4, and days until it results in the gelation when heating at 115 degrees C were shown. As compared with the conventional electrolytic solution, it turns out that it is very hard to gel the electrolytic solution of this invention.

[0034]

[Effect of the Invention] If the electrolytic solution of this invention is used, an electrolytic capacitor with more high rated voltage can be realized by low loss, and

industrial value is more large.

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[Translation done.]